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Development of precise nanosecond and picosecond dye lasers for spectroscopy of antiprotonic atoms

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Abstract Two laser systems were developed by the ASACUSA collaboration of CERN to carry out spectroscopy experiments on antiprotonic atoms. One of these was a continuous-wave pulse-amplified dye laser which was frequency-stabilized to a femtosecond frequency comb. The other generated 700-ps-long laser pulses of wavelengths $\lambda = 266$ and 532 nm using two stimulated Brillouin scattering cells filled with water.

Keywords Antiprotonic helium · Nanosecond dye laser · Frequency chirp

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1 Introduction

The ASACUSA collaboration of CERN has recently carried out several laser [1–4] and microwave [5] spectroscopy experiments on antiprotonic helium atoms ($\overline{p}\mathrm{He}^+ \equiv \overline{p} + \mathrm{He}^{2+} + e^-$) and ions [6]. In these experiments, laser transitions of wavelengths $\lambda = 264$–726 nm were induced between Rydberg states of the antiproton, with principal and angular momentum quantum numbers of $n \sim \ell - 1 \sim 40$.

Only nanosecond lasers can provide the megawatt-scale output energies needed to induce these antiproton transitions within the 1-μs-scale lifetime of the atom against antiproton annihilation in the helium nucleus. However, fluctuations in their

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frequency and linewidth, and the difficulty in calibrating their wavelengths generally limit the fractional precision attainable with these lasers to around 1 part in 10^6. Over the years we refined these nanosecond lasers, thereby measuring the $\overline{p}\mathrm{He}^+$ frequencies with successively higher precisions, from 2 parts in 10^7 [2] to 1 part in 10^8 [3, 4]. By comparing the results with three-body QED calculations [7–9], the antiproton-to-electron mass ratio was recently derived [4]. In this paper, we describe this developmental work.

2 Grating-stabilized pulsed dye lasers

The laser systems described here were all housed in a 10 m × 10 m clean room built inside the Antiproton Decelerator (AD) facility of CERN. The initial series of experiments [2] used a commercial dye laser (Coherent Scanmate-2E) pumped by the second or third harmonic of a Q-switched Nd:YAG laser (Fig. 1a). The output wavelengths $\lambda = 529$–745 nm were measured by four Fizeau interferometers (Cluster Lambdameter) embedded in a thermostabilized monolithic quartz block filled with neon gas [10]. These readings were calibrated by carrying out optogalvanic spectroscopy of neon and argon, or saturation spectroscopy of the rovibrational lines of molecular iodine and tellurium. The calibration accuracy was around 20–50 MHz.

The 30-cm-long dye laser cavity contained a Littrow-mounted grating and intracavity etalon of bandwidths $\Gamma \sim 6$ and 1.2 GHz. Three longitudinal cavity modes spaced by intervals of 0.5 GHz coexisted in the laser's gain envelope. The laser wavelength and longitudinal mode intensities fluctuated from pulse to pulse by more than 1 GHz. These quantum fluctuations arise because it is the first few photons, emitted spontaneously from the dye and proceeding to occupy the multiple cavity modes, that initiate the lasing process. We rejected [10] laser pulses that deviated from a single-mode structure, thereby effectively narrowing the linewidth of the laser to around $\Gamma = 0.4$–0.5 GHz.

The laser system was robust and its wavelength could be rapidly changed. Some ~ 20 transition frequencies of the $\overline{p}^4\mathrm{He}^+$ and $\overline{p}^3\mathrm{He}^+$ isotopes [2, 3], and the populations [11, 12] and Auger rates [13] of the associated states were systematically measured using the laser.

3 Continuous-wave pulse-amplified dye lasers

We later based our experiments [4] on single-line-mode continuous-wave (cw) dye and titanium sapphire (Ti:S) lasers of output power $P \sim 1$ W and linewidth $\Gamma = 0.1 - 1$ MHz (Fig. 1b). The optical frequencies of these lasers were stabilized against a femtosecond frequency comb [14, 15] with a precision of ~ 200 kHz. The comb constituted a mode-locked Ti:S laser pumped by a cw Nd:YVO_4 laser, which produced 15-fs-long laser pulses of repetition rate $f_r = 200$ MHz and average power $P = 0.7$ W. The cw laser was locked 20 MHz above the $n = 1592190$–2607811 th mode of the comb. The laser frequency was scanned by changing the repetition rate from 200.000 to 200.004 MHz.

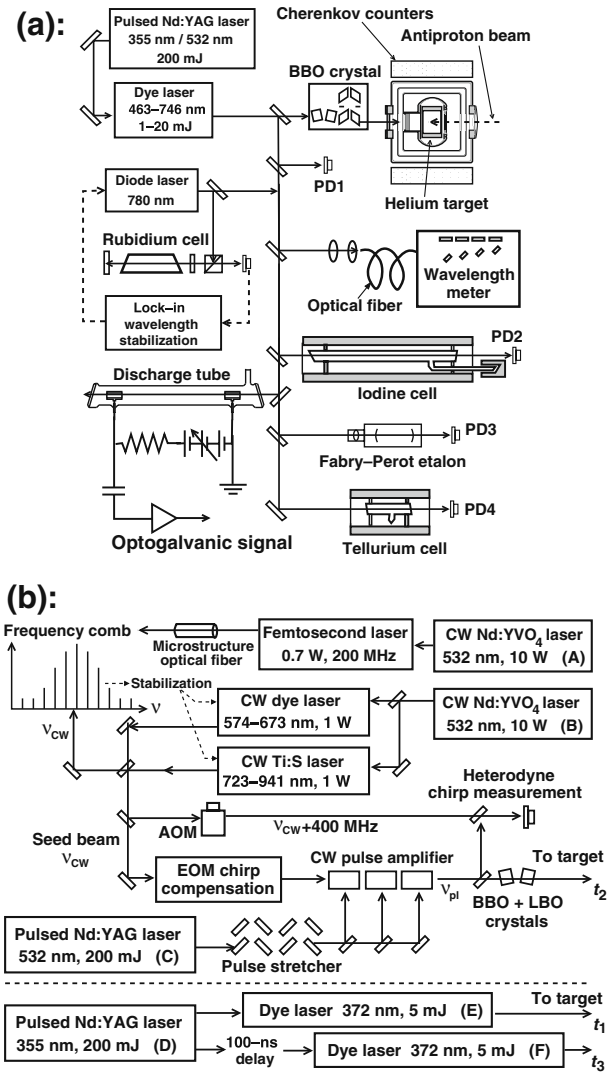

Fig. 1 Grating-stabilized (**a**) and cw-pulse amplified (**b**) dye laser systems used for spectroscopy experiments on antiprotonic helium, see text

The cw seed beam was amplified by factor $\sim 10^6$ in three dye cells pumped by the above Nd:YAG laser [16]. To decrease the Fourier-limited linewidth of the pulsed dye laser, the pump beam was split into seven beams, to which incremental delays were added. These were then merged to produce the stretched (20-ns-long) pulse that irradiated each cell. The energy and linewidth of the pulsed dye laser were $\Gamma = 60$ MHz and $E = 5 - 20$ mJ. UV wavelengths $\lambda = 264.7$–470.7 nm were obtained by either frequency doubling or tripling in beta-barium borate (BBO) or lithium triborate (LBO) crystals. The frequency of the dye laser pulse can deviate from the seed value by several tens MHz, due to sudden changes in the refractive index of the dye during the amplification. We corrected this so-called "frequency chirp" by using an electro-optic modulator to apply a frequency shift of opposite sign to the seed laser [16, 17].

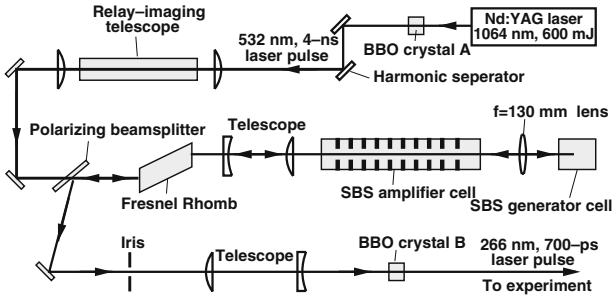

Fig. 2 Layout of Nd:YAG laser with SBS cells, which produced 700-ps-long laser pulses

4 Picosecond pump laser generated using SBS cells

Some of the experiments on antiprotonic helium ions [6] required short UV laser pulses of duration $\Delta t < 1$ ns. We used two stimulated Brillouin scattering (SBS) cells [18] to temporally compress the output of an injection-seeded Q-switched Nd:YAG laser (Fig. 2). The amplifier cell constituted a quartz tube of diameter $d = 40$ mm and length $l = 1.2$ m, with UV-transparent fused silica windows bonded on the two ends. Deionized water was continuously circulated through the cell. The leading edge of the laser pulse emerged from the other side of the SBS amplifier cell, and was focused into a second SBS generator cell filled with deionized water. This latter cell acted as a phase-conjugating mirror, so that the SBS Stokes beam emerged from the focus with the opposite phase and propagation direction from the original incident beam. A strong acoustic wave was excited in the amplifier cell, and successive sections of the incident laser beam reflected off the leading edge of this growing wave. The intensity of the SBS laser light therefore increased coherently as its pulse length was compressed. We obtained laser pulses of $\Delta t = 700$ ps duration and energy $E = 5$ mJ at the fourth harmonic of $\lambda = 266$ nm [19]. This beam was then used to pump the above dye lasers, as as Ti:sapphire pulsed lasers recently developed by us [20].

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